Environmental Research 162 (2018) 63–73

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

What's hot about mercury? Examining the influence of climate on mercury levels in Ontario top predator fishes

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ARTICLE INFO

Keywords:
Climate change
Predatory fishes
Fish mercury
Industrial emissions

ABSTRACT

Mercury (Hg) levels in Ontario top predator fishes have been increasing in recent decades. These increases may be a result of many additive factors, including global climate change. Only recently has research been conducted on how climate change may impact Hg levels in freshwater fishes at large-scales. We examined the relationship between Hg trends and (1) local weather, (2) large-scale climate drivers, and (3) anthropogenic Hg emissions, in native cool water (walleye and northern pike) and warm water (smallmouth bass and largemouth bass) predatory fishes in Ontario, Canada, for historical (1970–1992) and recent (1993–2014) time periods. For each fish species studied, > 25% of Ontario's secondary watersheds shifted from historically declining to recently increasing fish Hg trends, and ≥ 50% of watersheds experienced increasing trends between 1993 and 2014. Recent fish Hg increased at up to 0.20 μg/g/decade; which were significant ($p < 0.05$) for walleye, northern pike and smallmouth bass. Multiple linear regressions revealed a complex interplay of local weather, large-scale climate drivers, and anthropogenic Hg emissions influencing fish Hg levels. Recent Hg levels for walleye and largemouth bass increased with changes in global climate drivers, while higher precipitation influenced smallmouth bass Hg levels the most. Walleye Hg levels increased during the positive phases of global climate drivers, reflecting the local influence of local temperatures and precipitation indirectly. Differentiating the effects of climate-related parameters and emissions is increasingly crucial to assess how changing multiple environmental stressors may impact health of wildlife and humans consuming fish.

1. Introduction

Mercury (Hg) is a toxic heavy metal that can bioaccumulate and biomagnify in the food web, and adversely impact humans through consumption of fish (Mergler et al., 2007; Grimalt et al., 2010). By the 1970s, fish Hg levels in North America were substantially elevated due to industrial emissions, particularly those from coal-fired power plants (Downs et al., 1998). Although anthropogenic Hg emissions in North America have declined by approximately 75–90% between the 1970s and 2011 (Cain et al., 2007; Environment Canada, 2015), fish Hg levels are again increasing in the Province of Ontario, Canada (Monson et al., 2011; Tang et al., 2013; Gandhi et al., 2014). This mismatch in the trends of Hg emissions and fish levels suggests that other factors may be driving Hg dynamics.

Fish Hg levels can be influenced directly by factors such as lake size (Bodaly et al., 1993), lake acidity, hardness, dissolved organic carbon (DOC) (Wren et al., 1991), food chain length (Cabana et al., 1994; Pouilly et al., 2013; Johnson et al., 2015), trophic position (Coelho et al., 2013), species, size, sex (Gewurtz et al., 2011; Karimi et al., 2013), land use or land cover (Bank et al., 2006; Drenner et al., 2013), as well as indirectly by watershed disturbances such as forest fires and the invasion of non-native species in aquatic environments (Coelho et al., 2013; Dijkstra et al., 2013; Pack et al., 2014). The interactions of these factors can be complex, and become more complicated under climatic changes (IPCC, 2013). The impacts of climate factors, such as increased temperatures, changes in precipitation, wind patterns or dust deposition, can change the distribution, mobility and uptake of Hg in freshwater ecosystems in both direct and indirect pathways (Grimalt et al., 2010; Dijkstra et al., 2013; Evans et al., 2013; Eagles-Smith et al., 2017).

Deposition of atmospheric Hg can be a major contributor to Hg in fish (Pacyna et al., 2006; UNEP, 2013) and the amount deposited on to aquatic ecosystems can be affected by climatic factors such as precipitation (Outridge et al., 2008; Grimalt et al., 2010; Risch et al., 2012). Mercury is a global pollutant that can be transported long distances (Engstrom, 2007; Krabbenhoft and Sunderland, 2013). This long distance transport is reflected in the total anthropogenic Hg deposited in Canada as > 95% is currently from trans-boundary sources (Risk

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https://doi.org/10.1016/j.envres.2017.12.018
Received 29 August 2017; Received in revised form 16 November 2017; Accepted 20 December 2017
Available online 26 December 2017
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Management Strategy for Mercury, 2010). Precipitation is one of the primary pathways through which reactive Hg enters lakes and watersheds, and influences the transport and distribution of Hg between systems (Outridge et al., 2008; Grimalt et al., 2010; Risch et al., 2012; ICMGP, 2017). Greater quantities of precipitation can elevate wet Hg deposition by incorporating water-soluble inorganic Hg (Hg^{II}) into rain or snow, and thereby increasing inputs watersheds and lakes (Lamborg et al., 2002), exposing fish and other aquatic organisms to this contaminant.

Temperature is another climatic factor that has been shown to influence fish Hg concentrations. Temperature can affect the amount of bioavailable methylmercury (MeHg) at the base of the food chain as it is highly correlated with epilimnetic temperatures and it can also influence the rates of atmospheric transport between systems (Adrian et al., 2009; Grimalt et al., 2010; Pack et al., 2014). Available MeHg can be directly affected by temperature in a variety of ways. For example, warmer temperatures have been shown to increase the conversion rate of Hg^{II} to MeHg (Avramescu et al., 2011; Hecky et al., 1991; Bloom, 1992). In addition, warmer epilimnion temperatures have resulted in methylation rates exceeding demethylation by sulfur-reducing bacteria, one of the main converters of Hg^{II}, increasing the net MeHg available and exposure to aquatic organisms (Hecky et al., 1991). Elevated temperatures can also indirectly influence available MeHg in aquatic ecosystems by reducing dissolved oxygen (Jankowski et al., 2006). Mumley and Abu-Saba (2002) found that methylation of Hg quadrupled when dissolved oxygen concentration dropped below 6 mg/L. Lastly, global models have also projected that warmer temperatures may result in less oxidation, causing elemental mercury (Hg^0) levels to remain longer in the atmosphere, enhancing long-range transport of Hg (Wängberg et al., 2010). With rising temperatures due to climate change, we can expect higher methylmercury (MeHg) content in the food web (Bodaly et al., 1993; Canário et al., 2007; Stern et al., 2012).

The available Hg found in freshwater lakes can also be partially attributed to changes in global climate drivers. Fluctuations in air circulation patterns, or oscillations of large-scale climate drivers, have significant effects on three main factors: (1) local climate (Li et al., 2013; Zhao et al., 2013; Evans et al., 2013), (2) local weather patterns, and (3) the transport of Hg from distant sources to local lakes, especially those from Asian industrial locations (Selin et al., 2007). For example, negative phases of the El Niño Southern Oscillation (ENSO) are associated with warmer winter temperatures (George et al., 2000; Bonsal and Shabbar, 2011) and below average winter precipitation in Ontario (Bonsal and Shabbar, 2011). Changes in local air circulation, precipitation and temperature patterns attributed to large-scale climate drivers, can therefore indirectly affect fish Hg levels (Evans et al., 2013). Evans et al. (2013), for example, found that the Pacific/North American Index (PNA) was important in explaining Hg levels in lake trout in Great Slave Lake, Northwest Territories, Canada, indicating that trends may have been affected by patterns in air circulation.

Although most studies often attribute the increase in fish Hg levels to multiple factors, such as global Hg emissions or climate change (Tang et al., 2013; Gandhi et al., 2014; ICMGP, 2017), the proportional influence of climate on fish Hg levels is still unclear. We investigate the relative influence of local weather, large-scale climate drivers, and atmospheric Hg emissions on fish Hg levels in native cool water (walleye, *Sander viteus*, WE; northern pike, *Esox lucius*, NP) and non-native warm water (smallmouth bass, *Micropterus dolomieu,* SMB; largemouth bass, *Micropterus salmoides,* LMB) fishes in secondary watersheds of the Province of Ontario, Canada. Secondary watersheds are defined catchment basins of land, drained by a watercourse and its tributaries by the Ontario Ministry of Natural Resources. Secondary watersheds are subdivisions of the primary watersheds in Ontario and are either large river systems or groupings of small coastal streams. Specifically, the objectives of this study are two-fold: (1) to examine how Hg levels in Ontario's top predator fish have changed between historical and recent time periods at a secondary watershed level; and (2) to investigate how local weather, large-scale climate drivers, and local/global Hg emissions are potentially driving fish Hg trends over time. To our knowledge, this is one of few studies exploring the changes in fish Hg levels across a large landscape and interacting multiple environmental stressors, including climate change and atmospheric pollution. Since there is little climatic variation between lakes in the same region, we chose to examine fish Hg levels between watersheds in order to capture greater regional differences between watersheds. We predict that increases in fish Hg levels over time may more recently be attributed to changes in climate, specifically increases in precipitation events and temperature. This study is aimed to critically evaluate observed fish Hg trends under multiple stressors over broad spatio-temporal scales and improve our understanding of how intricate climatic processes can impact fish Hg levels and thereby health of fish, and the wildlife and humans consuming fish.

2. Materials and methods

2.1. Data acquisition and screening

Total fish Hg measurements were obtained from the Ontario Ministry of Environment and Climate Change (MOECC) Fish Contaminant Monitoring Program. This program was implemented to issue fish consumption advisories based on measured contaminants in Ontario fish. The program has collected Hg data since the 1970s from 2047 lakes, reservoirs, rivers, creeks and streams. Fish samples were collected in partnership with the Ontario Ministry and Natural Resources and Forestry (MNRF) during late summer or early fall using a variety of methods, including gill netting, trap netting, electrofishing, and angling. Total length, wet weight and sex (if possible) were recorded for each fish. Skinless boneless dorsal fillets were taken and stored at − 20 °C. Total mercury analysis using MOECC protocols were performed, including acid digestion and cold vapour flameless atomic absorption spectroscopy as described by Bhavsar et al. (2010) and Neff et al. (2012). We considered top predator fish because they often exhibit higher Hg levels than lower trophic level fish due to biomagnification (Kamman et al., 2005). Mercury data for four sport fish—WE, NP, SMB, and LMB—were screened for further analysis as detailed in the following section. We chose to include several predator species as different species experience Hg dynamics in distinct ways. By including more than one species we were able to infer fish biology differences inherent in food webs and gain a better understanding of the temporal trends in predator fish Hg levels (Bhavsar et al., 2010). For this study, Hg measurements for only natural inland lakes (excluding the Great Lakes) were retained, while reservoir, river, creek, and stream data were omitted. The final Hg dataset before further screening consisted of 36,639 WE measurements from 1232 locations; 25,978 NP measurements from 1313 locations; 11,879 SMB measurements from 652 locations; and 3340 LMB measurements from 217 locations sampled between 1970 and 2014.

It is well-known that fish Hg concentrations increase with fish size (Gewurtz et al., 2011). To reliably assess changes in fish Hg concentrations over time, Hg levels were first standardized at medium lengths for each species, using power series regressions (Supplementary Table A.1; Gandhi et al., 2014). A total of 6159 power series regressions, one for every combination of fish species, watershed, and year, were conducted (2033 WE, 1901 NP, 972 SMB and 300 LMB). Standard total lengths representing medium sizes were selected for each fish species based on previous literature (Scott and Crossman, 1998; Gewurtz et al., 2011; Gandhi et al., 2014). To avoid using over-extrapolated concentrations of Hg at the standard lengths, only fish sampled within 15 cm of the maximum and minimum medium lengths were considered. For example, to calculate the Hg concentration for a 40 cm WE at each location and year, only sampling events with the smallest WE length being no greater than 55 cm and the largest WE length no less than 25 cm were retained (Table S1). The final standard length

dataset for analysis consisted of 5272 Hg concentration data points for all species/length/location/year, for only medium sized fish (50 cm for WE, 60 cm for NP, and 30 cm for SMB and LMB).

Climate and emissions variables were obtained from multiple open access data sources (Table S2). Predictor variables were split into three main categories: (1) local weather, (2) large-scale climate drivers, and (3) Hg emissions. Local weather variables included mean monthly precipitation (Precip) and average daily temperature (Temp) from 1970 to 2013 and were obtained from the Climatic Research Unit (CRU, see http://www.cru.uea.ac.uk/). CRU provides monthly weather and precipitation data since 1900 at a spatial resolution of 0.5° latitude × 0.5° longitude interpolating over 4800 meteorological stations around the world (Harris et al., 2014). A total of 9 large-scale climate driver indices were also obtained: El Nino Southern Oscillation index (ENSO), North-Atlantic Oscillation index (NAO), North Pacific Oscillation index (NPO), Pacific Decadal Oscillation index (PDO), Polar/Eurasian pattern (PE), total Sunspot number (SunTOT), group Sunspot number (SunGN), Tropical/ Northern Hemisphere pattern (TNH), and West Pacific pattern (WP) from various sources (such as NOAA; see Table A2). These climate drivers were chosen for their predicted impact on the temperature, precipitation, air circulation, etc. within Ontario watersheds (Bonsal and Shabbar, 2011). Lastly, global and Canadian Hg emissions were acquired from Muntean et al. (2014), the European Monitoring and Evaluation Programme (EMEP, 2015) and Environment and Climate Change Canada (Environment Canada, 2015). Further details of climate variables can be found in the supplementary table (Table A2).

The purpose of our study was to examine the spatio-temporal relationships between climate and fish Hg levels at a landscape scale across all years between 1970 and 2013. Many previous studies have focused on fish Hg levels at the local scale, yet with the potential increasing influence of global emissions and the decline of point sources, assessing trends at broader spatial scales is necessary (Temme et al., 2007; Pacyna et al., 2010;). To justify this approach, non-parametric Kruskal-Wallis (KW) tests were performed in order to assess if fish Hg concentrations were significantly different among lakes within the same secondary watershed (see Fig. 1). The KW tests were conducted among standard length fish Hg levels of the same species within a 5-year time period (*e.g., lakes in secondary watershed A from 1970 to 1974)*. If fish Hg concentrations were not statistically different at a significance level of $p = 0.05$, they were merged to obtain watershed-specific standard length fish Hg concentrations to identify spatial and temporal trends. Only those cases with were more than 2 observations for each fish species/size/time period were considered. We found that for each species and standard length, fish Hg levels within a secondary watershed and 5-year time period were not significantly different ($p > 0.05$). As such, we determined it appropriate to conduct fish Hg trend analysis at the secondary watershed level. Data for the Ontario watersheds were obtained from the MNRF metadata website (see www.ontario.ca/data).

2.2. Data analysis

2.2.1. Temporal trend analysis

A nonparametric Sen's slope estimate assessed linear fish Hg trends for the historical (1970–1992) and recent (1993–2014) time periods for each secondary watershed. Sen's slope estimator is a conservative, non-parametric approach to estimating trends as it represents the median of the slopes calculated between each pair of points (Sen, 1968). These time periods were chosen because they capture the historical and recent years of fish Hg change in the Laurentian Great Lakes region and for Ontario's inland lakes; particularly in light of changes in policy regarding Hg point source pollution from industry. Further, these time periods were guided by results from Gandhi et al. (2014), in which they found significant increases in fish Hg levels between 1995 and 2012 for two Ontario predatory fish species. The recent time period encapsulates changes in fish Hg levels not associated to historical emission sources or point-pollution. The first time period (1970–1992) includes years where this policy was newly implemented, while recent years (1993–2015) represents a time that should have greatly reduced point sources contributing to fish Hg levels (Bhavsar et al., 2010; Gandhi et al., 2014). Sen's slopes were calculated using the MAKESENS (Mann-Kendall test for trend and Sen's slope estimates) Microsoft Excel Template (Salmi et al., 2002).

2.3. Correlation analysis with climate and emissions variables

Spearman's rank correlations were used to quantify the relationship between fish Hg levels and all climate and Hg emission variables for the historical, recent and 1-year lag time-periods for each species and watershed. We included a lag of 1 year for all weather, climate, and emissions variables as it has been suggested that bioaccumulation of Hg to reach top predator fish ranges from 1 to 3 years (Harris et al., 2007). Additional time lags up to 3 years were tested, but the effects were not significant (Chen, 2015). To ensure that correlations between fish Hg and climate variables were not masked by trends in the local climate data, residuals of fish Hg and local climate variables (mean daily temperature and monthly precipitation) were used. A Holm-corrected Sequential Bonferroni (Holm, 1979) procedure was also conducted to control for family wide statistical error. Altogether, over 1400 correlation analyses were conducted between predictor variables and fish Hg levels for each species in each watershed per time period and lag.

2.4. Modelling Ontario fish Hg levels with climate and emissions variables

Multiple linear regression models for each fish species were developed across Ontario secondary watersheds. As the literature suggests a lag time in the bioaccumulation of Hg in top predator fish, we used 1-year lagged fish Hg concentrations in the regressions (Harris et al., 2007). Fish Hg concentrations were assessed for normality using a Shapiro-Wilks test ($p < 0.05$) and log-transformed to meet the assumptions of normality before developing multiple linear regressions. Fish Hg and local climate variables were detrended and residuals were used in place of raw data to develop the regression. A forward selection procedure with a dual-criterion, such that to be included in the model, predictor variables were significant at $\alpha = 0.05$ and explained significant additional variation (R^2_{adj}), was used to identify significant weather, climate, and emissions predictor variables for fish Hg levels in WE, NP, SMB and LMB (Blanchet et al., 2008). There was no limit on the number of predictor variables to include in the model.

We conducted a Redundancy Analysis (RDA) to identify associations between average fish Hg levels and local weather, large-scale climate drivers, and anthropogenic Hg emissions predictor variables in one south-east watershed of Ontario where all fish species were present. Though we were only able to develop an RDA for one watershed, results from the RDA would help to guide linear model building. Only 15 years of Hg data for all four fish from within the watershed were included in the RDA because of missing Hg data. Average fish Hg data for WE, NP, SMB, and LMB were included as the response variables for the RDA (represented by points). Local weather, large-scale climate driver, and emissions, in addition to year (represented by arrows) were the explanatory variables used to describe the variation in fish Hg levels in a south-western Ontario secondary watershed. Analyses were performed in Microsoft Excel (Microsoft Office, 2010, Version 14.0), using the *glm, forward.sel, and rda* functions in the MASS, packfor and vegan packages in the R-language environment (R Development Core Team, 2015; version 3.2.1, MASS package) and mapped using ArcGIS software (ESRI, 2015). The data acquisition and analysis framework is summarized in Fig. 1.

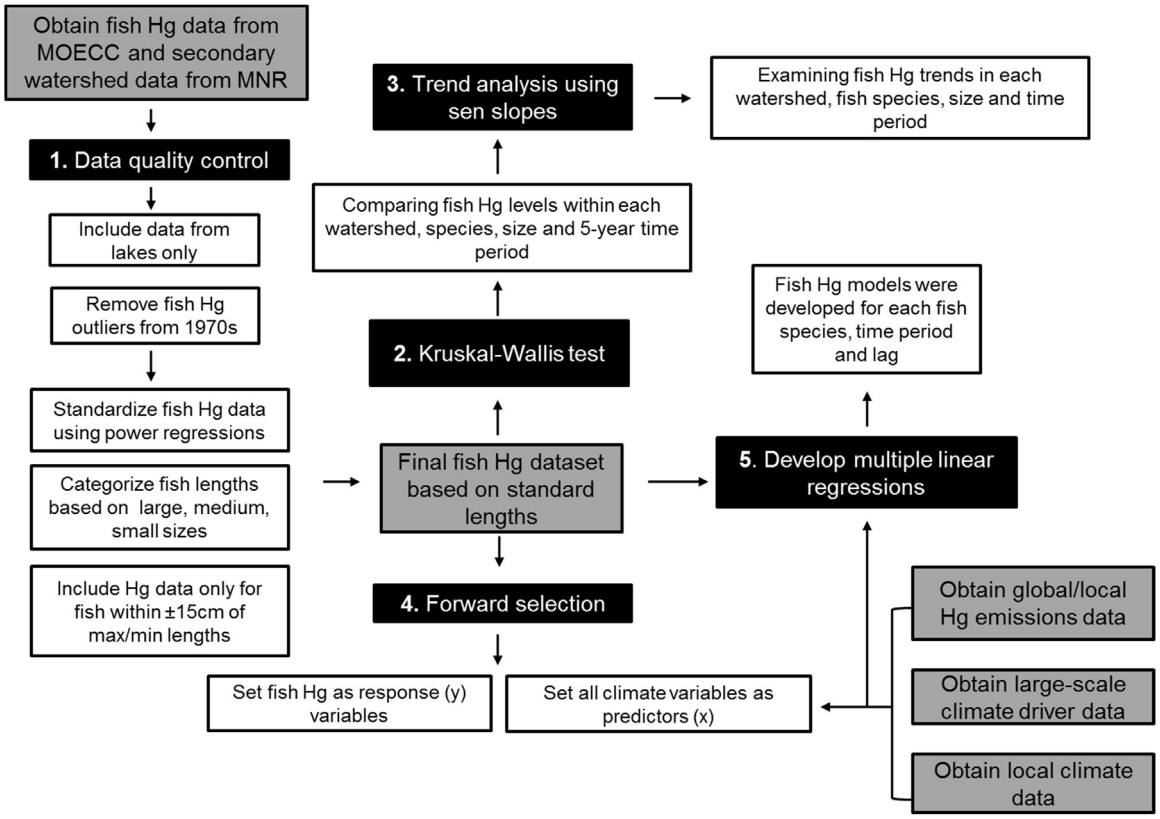
Obtain fish Hg data from MOECC and secondary watershed data from MNR
1. Data quality control
Include data from lakes only
Remove fish Hg outliers from 1970s
Standardize fish Hg data using power regressions
Categorize fish lengths based on large, medium, small sizes
Include Hg data only for fish within ±15cm of max/min lengths
Final fish Hg dataset based on standard lengths
4. Forward selection
Set fish Hg as response (y) variables
Set all climate variables as predictors (x)
5. Develop multiple linear regressions
Obtain global/local Hg emissions data
Obtain large-scale climate driver data
Obtain local climate data
Fish Hg models were developed for each fish species, time period and lag
2. Kruskal-Wallis test
Comparing fish Hg levels within each watershed, species, size and 5-year time period
3. Trend analysis using sen slopes
Examining fish Hg trends in each watershed, fish species, size and time period

Fig. 1. Method framework. Grey boxes indicate data or datasets; black boxes indicate data screening or analyses; and white boxes provide detail on the data analysis. Refer the Methods section for details.

3. Results

3.1. Historical and recent fish Hg trends

Comparing fish Hg levels across Ontario secondary watersheds, we find that a majority of watersheds do not have significant changes in fish Hg concentrations. However, we have identified watersheds in Ontario that have significantly decreased in fish Hg historically, and now increased in recent time periods. Fish Hg levels for two cool water predators (WE and NP) largely decreased during the historical period (Fig. 2a,c), but mostly increased in recent decades (Fig. 2b, d). For WE, between 1970 and 1992, 68% of the secondary watersheds experienced declines in fish Hg. Hg levels in WE in south-western Ontario decreased significantly ($p < 0.05$) at rates of 0.20–0.70 μg/g/decade (Fig. 2a). In contrast, for the recent time period (1993–2014), 57% of the secondary watersheds experienced increases in fish Hg. Recent Hg levels in northwestern Ontario WE increased at rates of 0.20–0.40 μg/g/decade, whereas historically Hg levels were significantly decreasing at a rate of 0.20–0.70 μg/g/decade. Between the time periods, 26% of the watersheds switched from decreasing to increasing Hg levels, whereas 4% of watersheds switched from increasing to decreasing.

Similarly, NP Hg historically declined in 71% of the watersheds with non-significant ($p < 0.05$) declines in northern, western, and east-central Ontario at rates of 0.10–0.70 μg/g/decade (Fig. 2c). In the recent time period, 52% of the secondary watersheds showed increasing NP Hg levels, with NP Hg levels in western Ontario increasing at 0.20–0.30 μg/g/decade. About 32% of the watersheds switched from historically decreasing to recently increasing NP Hg levels and only 8% of watersheds changed from increasing to decreasing.

The warm water predatory fishes (SMB and LMB) are currently restricted to southern and south-central Ontario (Fig. 2e,g), but have been expanding their range northwards during the time period of this study in response to warming temperatures (Alofs et al., 2014). Historically, SMB Hg concentrations either remain unchanged or decreased (at the rate as high as 0.20 μg/g/decade), whereas LMB Hg levels generally increased, albeit insignificantly (as much as 0.10 μg/g/decade; Fig. 2g). Between 1993 and 2014, SMB Hg levels increased in 53% of the watersheds at up to 0.20 μg/g/decade (Fig. 2f). Similarly, LMB Hg levels increased in 50% of the watersheds at up to 0.20 μg/g/decade (Fig. 2h). For SMB and LMB Hg levels in the recent time period, approximately 38% and 30% of the watersheds switched from decreasing to increasing, and 7% and 25% of watersheds switched from increasing to decreasing, respectively.

3.2. Correlations for 1-year lag fish Hg and climate/emission variables

Spearman correlations of fish Hg and climate and emission variables at a lag of 1-year suggested that a single variable acting alone has not been primarily responsible for the patterns in fish Hg levels, but rather a complex interplay among local weather, large-scale climate drivers, and emissions is driving the fish Hg levels (Fig. 3). Cross-correlations for each fish species across watersheds revealed that generally large-scale climate indices have weak, non-significant correlations ($r = \pm 0.1$) over time. After controlling for family wide statistical error from multiple comparisons, correlations remained insignificant. However, correlations against local weather, large-scale climate, and emissions correlations data did vary for each fish species.

For example, historical WE Hg levels (Fig. 3a) were correlated with ENSO ($r = 0.3$), global ($r = -0.3$) and Canadian Hg emissions ($r = 0.3$), although in recent decades these relationships are closer to zero. The strongest correlations historically for NP Hg concentrations (Fig. 3b) were with TNH patterns ($r = -0.4$), global ($r = -0.3$) and Canadian Hg emissions ($r = 0.3$); while in recent time periods, average monthly precipitation ($r = 0.3$) held the strongest median correlation among variables. Historical SMB Hg levels (Fig. 3c) were correlated with global ($r = -0.4$) and Canadian Hg emissions ($r = 0.45$); while

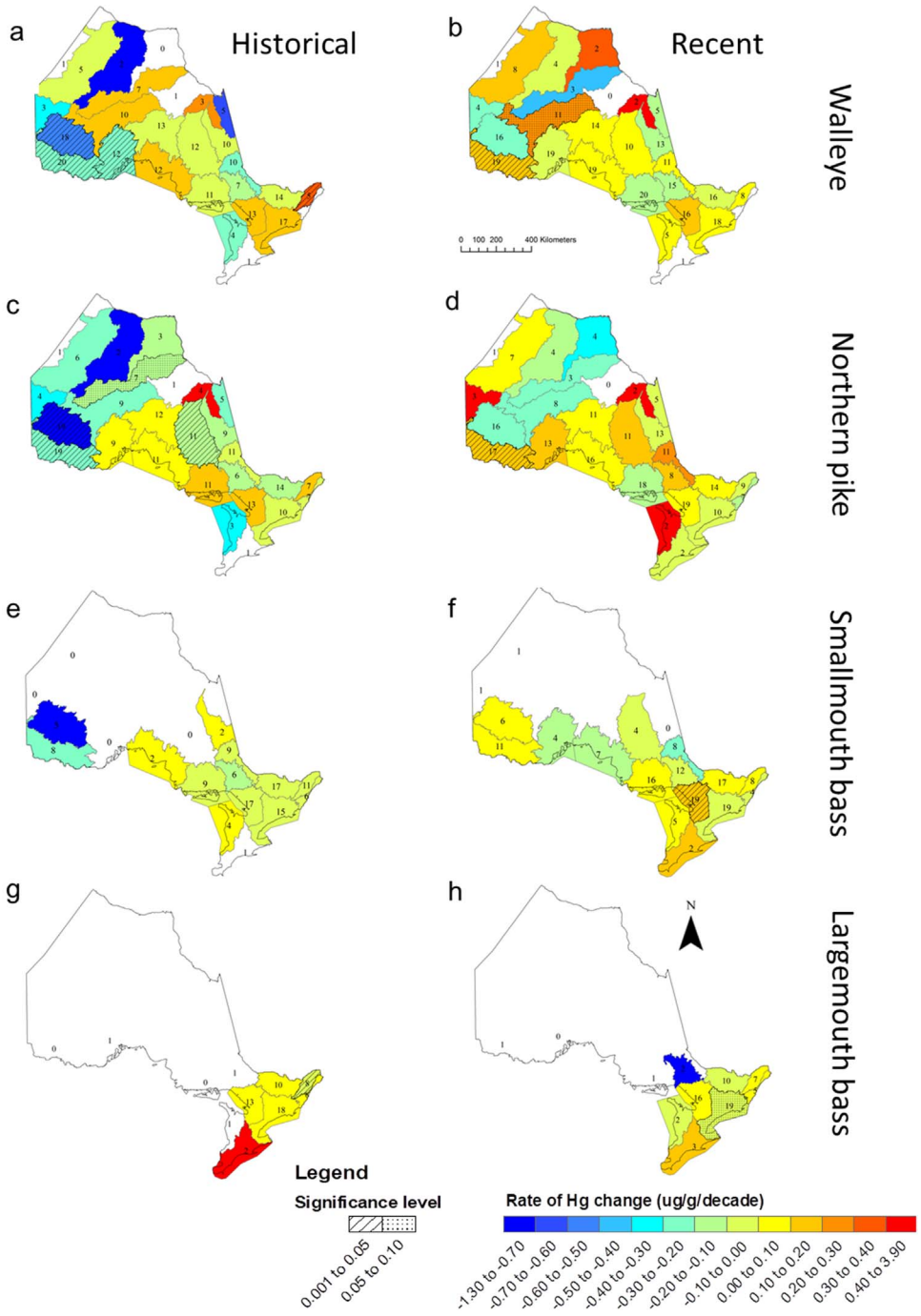

Fig. 2. Fish Hg trends for walleye (a, b), northern pike (c, d), smallmouth bass (e, f), and largemouth bass (g, h) in each secondary watershed for the historical (1970–1992) and recent (1993–2014) periods estimated using Sen's Slope. The number for each watershed represents the number of lakes. Extreme values are binned into the upper and lower ends of the scale so the variation is not lost. White watersheds indicate insufficient data for the trend analysis.

global emissions (r = 0.3) and average daily temperature (r = − 0.3) were most correlated against recent SMB Hg levels. Lastly, historical LMB Hg levels (Fig. 3d) had the strongest correlations with average monthly precipitation (r = 0.45), average daily temperature (r = − 0.5), and ENSO (r = 0.4). Recent LMB Hg levels correlated the most with average monthly precipitation (r = 0.3).

3.3. Explanatory models for lag 1-year fish Hg levels

Eighty-eight percent of the variation in the fish Hg levels of a southwestern Ontario secondary watershed where all fish species in this study are present, can be explained by local weather, large-scale climate drivers, and anthropogenic Hg emissions variables, based on an RDA. The ordination suggests that WE Hg levels are higher in warm years and positive phases of NAO. NP Hg levels appear to be higher during positive phases of NP and negative phases of PDO. For SMB, higher Hg levels are associated with wetter and, surprisingly, cooler temperatures. Lastly, LMB Hg levels are higher in positive phases of ENSO and negative phases of PE (Fig. 4).

Multiple linear regressions for historical and recent periods were developed to investigate the relationship between fish Hg levels in the four top predator freshwater fishes and climate/emission variables. Generally, recent fish Hg levels have increased with positive phases of large-scale climate drivers, and greater precipitation (Table 1).

Historically for WE, Hg levels increased during the positive phase of ENSO and explained 9% of the variation. In recent decades, WE Hg levels increased during the positive phases of NPO and WP, which explained 42% of the variation (Table 1). There were no significant

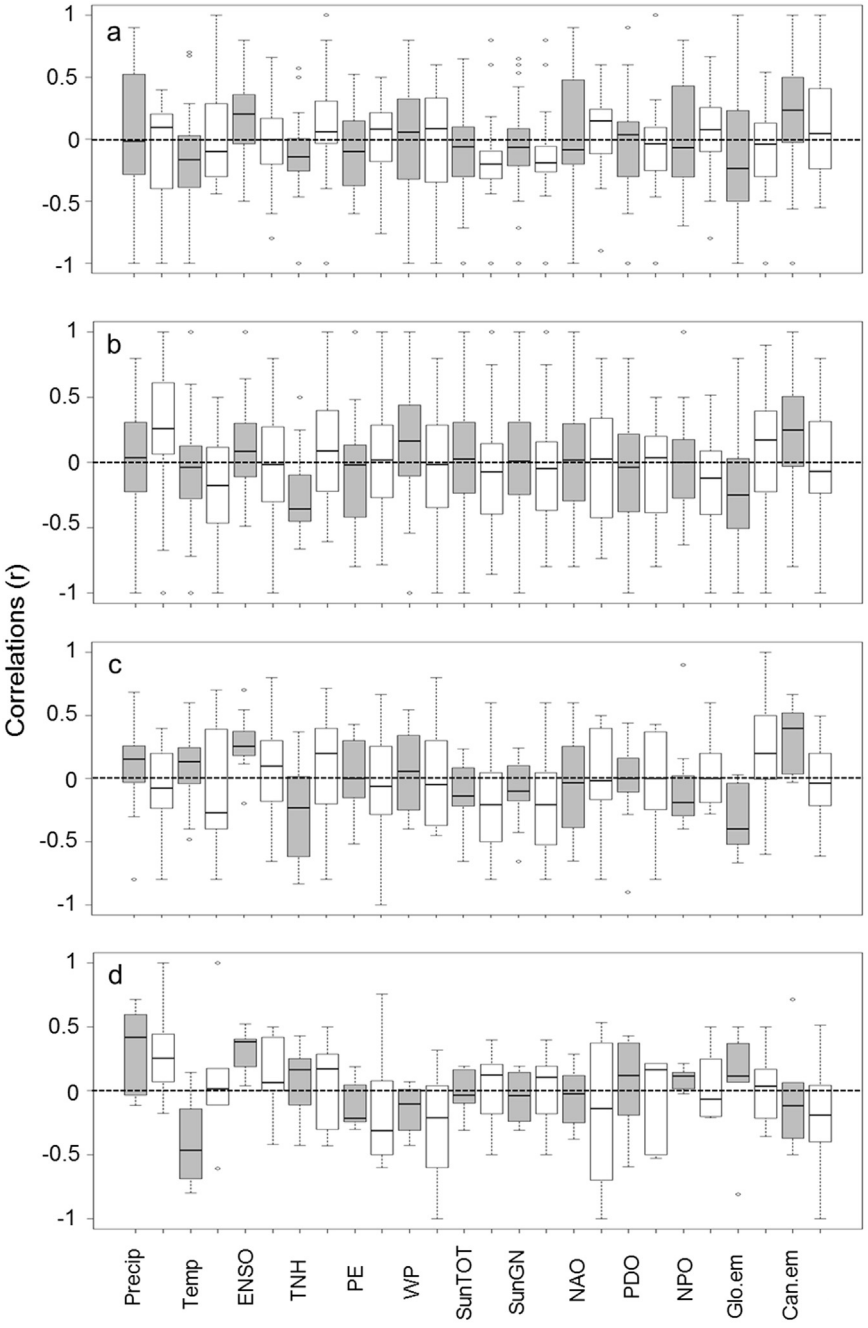

Fig. 3. Boxplots of correlations between 1-year lag fish Hg levels and climate/emission variables for the historical (1970–1992; grey) and recent (1993–2014; white) periods for (a) ye (572 correlations), (b) northern pike (598 correlations), (c) smallmouth bass (286 correlations) and (d) largemouth bass (130 correlations). Precip: average monthly precipitation; Temp: mean daily temperature; ENSO: El Nino Southern Oscillation index; TNH: Tropical/ Northern Hemisphere pattern; PE: Polar/Eurasian pattern; WP: West Pacific pattern; SunTOT: total Sunspot number; SumGN: group Sunspot number; NAO: North Atlantic Oscillation index; PDO: Pacific Decadal Oscillation index; NPO: North Pacific Oscillation index; Glo.em: Global Hg Emissions; Can.em: Canadian Hg Emissions.

variables explaining NP Hg levels historically or recently (Table 1).

ENSO was the most important predictor of SMB Hg concentrations historically and explained 11% of the variation. Precipitation explained 31% of the variation in SMB Hg levels in recent decades. Lastly, mean air temperatures explained 41% of the variation in the historical LMB Hg levels, whereas mean precipitation explained 10% of the variation in recent LMB Hg levels (Table 1).

4. Discussion

4.1. Trends

By examining fish Hg changes at the broader spatial scale of secondary watersheds, our study corroborated previous reports that Hg levels in Ontario's predatory fishes appear to have mostly decreased during the historic period of 1970–1992 and increased in the past two decades, although trends were not statistically significant. Further data collection and effort would be required to obtain statistically significant trends, however the overall direction of fish Hg trends between the two time periods are evident.

These changes in fish Hg levels over time also do not appear to be homogenous across the landscape of Ontario. For example, northwestern Ontario has shown more extensive decreases in fish Hg level historically compared to other regions. This is in response to the highly polluted conditions of lakes found in this watershed, including Wabigoon Lake, Clay Lake, Tetu Lake, Ball Lake, and Separation Lake (Neff et al., 2012), prior to the implementation of stricter policies on industrial emissions. From the late 1800s to early 1900s, small local gold mines near Dryden and Kenora may have contributed to elevated levels of Hg found in this area until 1920, in which mercury amalgamation was phased-out (Bruce, 1925; Pirrone et al., 1998). In later decades (60 s and 70 s), a chlor-alkali plant in Dryden Ontario

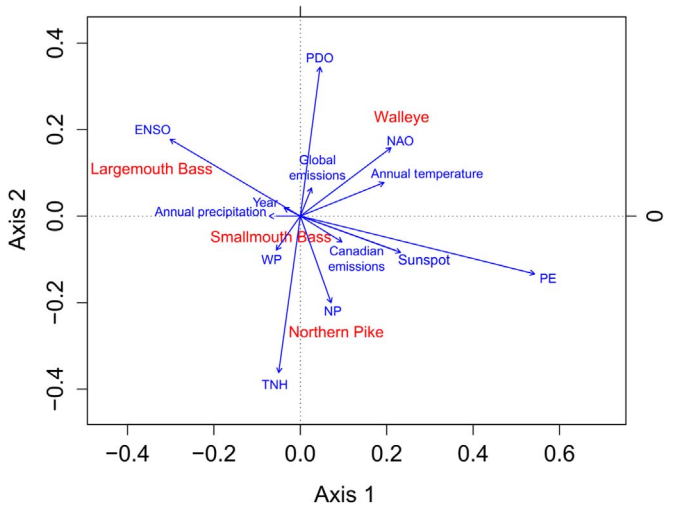

Fig. 4. Redundancy analysis (RDA) describing the association between fish Hg levels and weather, climate, and emission variables in a south-east Ontario secondary watershed where all fishes are present. Fish species are represented by points. Local weather, large-scale climate drivers, anthropogenic Hg emission variables, and years, are represented by arrows. The length of the arrow represents the strength of the relationship and the angle between arrows depicts the correlation between predictor variables. When a fish point is oriented close to the pointed end of an arrow, it suggests that fish Hg levels are associated to high values of that predictor variable. When the fish point is found 180° opposite of an arrow, it suggests that the fish Hg levels are associated to low values of that predictor variable. Weather, climate, and emission predictor variables describe 88% of the variation in fish mercury levels in a southern Ontario watershed (R^2 = 0.88).

Table 1
Multiple linear regression models relating 1-year lag Hg levels in four top predator freshwater fish and climate variables in historical (1970–1992) **and recent** (1993–2014) **time periods.** Walleye (WE) and northern pike (NP) represent cool-water predators, while smallmouth bass (SMB) and largemouth bass (LMB) represent warm-water predators. Local climate, emissions and large-scale climate indices are significant ($p \leq 0.05$) in predicting fish Hg levels.

Species	Time period	Predictor variables	Coefficient	R^2adj	p-value
WE	Historical	ENSO	0.006	0.09	0.1
NP	Historical	No variables			
SMB	Historical	ENSO	0.007	0.11	0.08
LMB	Historical	Mean air temperature	− 0.11	0.41	0.004
WE	Recent	NPO	0.03	0.16	0.04
		WP	0.1	0.26	0.08
NP	Recent	No variables			
SMB	Recent	Mean precipitation	0.01	0.31	0.01
LMB	Recent	Mean precipitation	0.005	0.1	0.1

(Armstrong and Hamilton, 1973) was an important source of Hg emissions resulting in a highly Hg contaminated area. Regionally, large-scale industrial emissions from the Great Lakes area may be linked to the level of Hg contamination in northwestern lake sediments (Wiklund et al., 2017; Pirrone et al., 1998). Anthropogenic Hg in lake sediment reached a maximum during the 1950s for lakes in the northwestern area, likely a reflection of the acute increases in Hg emission in the Great Lakes region and an increase in total North American emissions in the decade earlier (Wiklund et al., 2017). The decreasing trends in northwestern Ontario are more prominent between 1970 and 1993 as local and regional emissions were reduced during this time and the impact of this change was greater than in other watersheds.

4.2. Weather and climate

We found that changes in both local weather and large-scale climate may be contributing to recent increases in Hg levels in coolwater and warmwater predators. Between 1993 and 2014, fish Hg levels have increased at rates as high as 0.2 μg/g/decade for secondary watersheds in Ontario. These results are consistent with other studies on this topic (e.g., Monson et al., 2011; Tang et al., 2013; Gandhi et al., 2014). Gandhi et al. (2014) reported that Ontario NP and WE Hg levels increased by 0.01–0.27 μg/g/decade between 1995 and 2012. For boreal shield lakes in northern Ontario, Tang et al. (2013) found significant increases in rates of Hg bioaccumulation for WE and mean Hg concentration for NP between historical (1974–1981) and recent (2005–2010) time periods. Studies conducted for WE, LMB and NP in the Great Lakes regions demonstrated nonlinear Hg trends, with shifting upward Hg patterns in the early 1990s (Monson et al., 2011). These studies often attribute the increasing fish Hg trends to factors, such as rising global Hg emissions and climate change. Our study further investigates these claims to identify the role of weather, climate, and Hg emissions in driving fish Hg patterns over broad spatial and temporal scales.

Our analysis suggests that climate factors have become more influential on fish Hg levels in recent years. In historical time periods, climate variables (ENSO and mean daily temperature) were significant predictors of fish Hg levels. Similarly, within recent time periods, large-scale climate drivers (NPO and WP) and local climate (mean monthly precipitation) remained important predictors of fish Hg levels, but held greater explanatory power than historically. These models were informed by examining the associations depicted in ordination space. The clustering of fish Hg levels and explanatory variables in the RDA revealed some interesting associations, although the analysis was conducted on only one watershed over 15 years. Even with a smaller subset of data however, we found supporting evidence for the associations between large-scale climate drivers and WE, NP and LMB; and local weather variables for SMB.

4.3. Local weather conditions

Local weather was significant for both historical and recent fish Hg models, particularly for LMB and SMB. Interestingly, we observed a negative relationship between temperature and historical LMB Hg concentrations. This was also evident as a negative correlation with SMB and LMB Hg levels in recent time periods. We expected a positive correlation as freshwater fishes subjected to warmer waters often have increased metabolism (Bodaly et al., 1993; Canário et al., 2007; Pörtner and Knust, 2007; Wang and Overgaard, 2007; Stern et al., 2012) and several studies have shown that the increase in dietary intake can lead to increased fish Hg levels (MacCrimmon et al., 1983; Simoneau et al., 2005; Karimi et al., 2007). Since Hg is bioaccumulative, fish size and age are typically positively correlated with Hg level (Gewurtz et al., 2011). However, the negative relationship between temperature and fish Hg concentrations found in this study may be possibly attributed to growth dilution where at a given age, faster growing fish have generally lower Hg concentrations (Simoneau et al., 2005; Karimi et al., 2007).

It is also possible that a direct correlation of the individual variables with “net” trend may not be very informative and not fully encompass the effect of temperature on fish Hg concentrations. One possibility is that the magnitude of these correlations between temperature and fish Hg may be less negative than the correlations in the absence of a warming climate. In addition, the effect of temperature may have been positive, but large historical declines in the regional emissions were pulling the overall trend downward. Therefore, the negative relationship between temperature and fish Hg concentrations may be a product of using “net” trends to describe the relationship.

In addition to temperature, higher mean monthly precipitation translated to increased Hg levels in SMB and LMB models for recent time periods. This relationship was supported by our predictions based on the literature. Increased precipitation can facilitate increased Hg loading to aquatic environments through (1) re-mobilization via greater runoff from a watershed as a result of soil erosion, and (2) direct deposition (Risch et al., 2012; Wiener et al., 2012). Therefore, with a greater amount of Hg entering aquatic ecosystems due to increases in

precipitation, Hg concentrations in the food web and ultimately in top predator fish are also expected to increase.

4.4. Large-scale climate drivers

Fish Hg levels were higher typically during the positive phases of global climate drivers for the historical period. These climate indices are important in regulating local weather patterns, and thereby the environmental conditions to which fish are exposed (Forchhammer and Post, 2004; Shabbar, 2006; Bonsal and Shabbar, 2011). In this study, coolwater WE and warmwater SMB Hg levels were positively related with the ENSO Index. ENSO is one of the greatest drivers of inter-annual variation in Canadian climate (Bonsal and Shabbar, 2011). ENSO is known to influence the winter temperatures and total precipitation in Ontario, particularly the Great Lakes regions, with positive phases (La Niña) associated with above average precipitation and cooler temperatures (Shabbar and Khandekar, 1996; Bonsal and Shabbar, 2011; Yu et al., 2015). For example, French et al. (2006) identified that oscillations of total Hg concentrations in chinook salmon from the Bay of Quinte were associated with cooling La Niña trajectories and thus summer air temperatures.

In the recent time period, WE Hg levels increased with climate indices, NPO and WP. The positive phase of both the NPO and WP indices are associated with higher winter surface air temperatures in south-central regions of Canada including across the province of Ontario (Linkin and Nigam, 2008). Warmer winter temperatures can induce earlier ice break-up, decrease ice coverage and possibly result in the premature onset of lake stratification (Assel and Robertson, 1995; Hodgkins et al., 2002; Austin and Colman, 2007). These changes could subsequently lead to higher surface water temperatures, thus increases in methylation rates and decreases in demethylation rates (Hecky et al., 1991; Austin and Colman, 2007). The influence of large-scale climate drivers on local weather, such as increasing precipitation and temperature, generally positively impacts fish Hg trends in coolwater and warmwater predatory fishes.

Interestingly, for WE and SMB, our models suggested that large-scale climate indices were important predictors of fish Hg over local temperature and precipitation. Large-scale indices of climate are considered excellent predictors of ecological variation—even outperforming local weather variables because they can capture information on temperature, precipitation, and wind speed together and therefore represent a kind of “catch-all” climate variables for large scales (Hallett et al., 2004; French et al., 2006; Evans et al., 2013). For example, the NAO index encompasses anomalies in temperature and precipitation and also accounts for the changes in the strength of the westerlies. Changes in magnitude of the westerlies can affect winter storm frequency and severity (Rogers, 1984; Otterson et al., 2001) and possibly influence long-range transport of atmospheric Hg. For some indices, deviations of temperature and precipitation values during the same phase may not occur in the same direction. This is observed for ENSO in which the positive phase results in increases precipitation and decreases in temperature compared to average values (Shabbar and Khandekar, 1996; Bonsal and Shabbar, 2011; Yu et al., 2015). Furthermore, some climate indices may be associated with others affecting the strength of the phases and the magnitude of climate anomalies related to phases (ex. Barnston et al., 1991; Mo and Livezy, 1986). Therefore, indices incorporate many complexities that are not encompassed in local air temperature and precipitation and may be more suitable predictors of Hg in fish since the relationship between atmospheric Hg and fish Hg levels is also highly complex.

4.5. Anthropogenic Hg emissions

Over the last few decades, the U.S., Canadian and Ontario Hg emissions have declined due to stricter government regulations (Environment Canada, 2015). Although these regulations were expected to continue fish Hg declines, fish Hg levels appear to be mostly increasing in our study during the recent time period. The recent slowdown or reversal of decreasing fish Hg trends has been attributed to increases in global Hg emissions previously estimated (e.g., Gandhi et al., 2014; Pacyna et al., 2010). Surprisingly, our results suggest that anthropogenic Hg emissions did not provide any significant explanatory power within the fish Hg models. A recent study suggested that actions taken over the last two decades have resulted in lower global anthropogenic mercury emissions as well as deposition (Zhang et al., 2016). This may indicate that at the watershed scale, climatic variables may have a larger impact on fish Hg level than the current emissions themselves, and may be contributing to re-emission of past Hg pollution.

Beginning with Industrialization, Ontario's Hg problem was attributed to historically elevated emissions during the last 100–150 years. Historical emissions would have resulted in elevated deposition in the past. Today, the watershed repository is likely a more important contributor than the current emissions contributing to the present day deposition, since only 27% of global Hg deposition are from present human activities, while 13% are from natural source and 60% from re-emission of previously emitted and deposited mercury, also known as “legacy” Hg (Amos et al., 2013; Obrist et al., 2017). This re-cycling of previous mercury pollution may be exacerbated by changes in climate and may explain why our results indicate that Canadian and global Hg emissions are not prominent drivers of fish Hg levels. Therefore, despite decreasing Hg emissions from industry, if climate variables are driving increased Hg availability in lakes, it would be reflected in increasing fish Hg levels.

4.6. Landscape and local watershed characteristics

In addition to local weather and larger-scale climate drivers, regional landscape characteristics have also been shown to influence fish Hg levels. This may account for the variation in fish Hg trends in different watersheds as well as explain why certain watersheds were influenced by specific large-scale climate drivers and others were not. Land cover could possibly also help exacerbate or ameliorate the effects of climate change on fish Hg levels. In forested watersheds, the canopy effectively collects atmospheric Hg and via litterfall or throughfall can elevate the quantity of Hg transported to waterbodies (Rea et al., 1996; Graydon et al., 2008; Drenner et al., 2013), which can be enhanced by increases in precipitation. Specifically, Drenner et al. (2013) found that the percentage of coniferous forest coverage explained the most variation in largemouth bass and equivalent species (LMBE). LMBE Hg concentrations in 9 different ecoregions varied as much as 2-fold despite receiving similar amounts of wet deposition in which higher concentrations of Hg in fish were associated with greater coniferous forest coverage (Drenner et al., 2013). Furthermore, wetlands provide abundant organic matter and anoxic sediment for microbial methylation of Hg to form Me (Wiener et al., 2006; St. Louis et al., 1994). Through surface runoff or connected tributaries elevated MeHg levels [CH_3Hg^+] can be transported from watershed wetlands to nearby waterbodies and result in an increase in fish Hg levels. Agricultural lands may have the reverse effect on fish Hg levels compared to forests and wetlands. Phosphorus loading associated with agricultural activities can lead to algal bloom dilution and decrease the bioaccumulation of Hg in the food web (Pickhardt et al., 2002). In addition, this increased productivity can result in faster fish growth rates and reduce the bioaccumulation of Hg in fish known as growth dilution (Essington and Houser, 2003).

Implications of climate change on fish Hg levels may also go beyond changing precipitation patterns or climate drivers. A recent synthesis from the International Conference on Mercury as a Global Pollutant (ICMGP) focused on the key roles of extrinsic and intrinsic drivers that affect the exposure to and toxicity of Hg to humans and other animals. They named drivers such as climate change and invasive species

(Eagles-Smith et al., 2017), which are predicted to impact bioaccumulation of Hg, by altering food web structure and pathways of energy/biomass flow. Warming climate may facilitate northern invasions of warmwater fishes, including SMB and LMB. The introduction of an invasive species may alter trophic structure and lengthen the food chain, thereby impacting the levels of Hg in top predators (Hrabik et al., 1998; MacIssac, 1996; Vander Zanden and Rasmussen, 1996). The lengthening of food chains has been positively correlated with increased bioaccumulation of toxic contaminants in fish (Cabana et al., 1994; Vander Zanden and Rasmussen, 1996). For example, Rennie et al. (2010) proposed that the establishment of an invasive invertebrate predator species *Bythotrephes* in Ontario inland lakes lengthens aquatic food chains and thus increased fish Hg concentrations. Other than trophic position, variation in Hg levels between species has been attributed to diet and mobility (Harris and Bodaly, 1998; Kannan et al., 1998; Chumchal and Hambright, 2009). Predator consumption of pelagic prey particularly has found to increase levels of Hg in fish (Cabana et al., 1994; Power et al., 2002; Kidd et al., 2003). Thus with the introduction of SMB and LMB in novel environments in response to climate change, and the subsequent shift of native predators to dependence on pelagic food resources, Hg levels of SMB may potentially increase (Vander Zanden et al., 2004).

5. Future directions

Fish mercury concentrations have been increasing in recent years and the factors driving these positive trends such as changes in climate may continue over the next few decades. Though global emissions of Hg may increase in the future, there is a possibility of decreasing emissions with implementation of advanced emission control technologies especially in developing countries (Streets et al., 2009; Pacyna et al., 2010). Temperatures are projected to become warmer with less precipitation in Ontario (Bruce et al., 2003; Lemmen et al., 2008). Less precipitation may result in a decline in the wet deposition or the transport of Hg via runoff to aquatic ecosystems and increases in temperatures can result in growth dilution (Karimi et al., 2007; Risch et al., 2012; Wiener et al., 2012). However, it is likely that warmer temperatures may also increase methylation rates and invasion of warmwater species potentially leading to greater bioaccumulation of Hg in top predator fish (Hrabik et al., 1998; Bodaly et al., 1993; Stern et al., 2012). If global emissions and temperatures continue to increase this may lead to a continuation of the increases in fish Hg levels, which can have serious implications for the health of fish and people consuming these fish (Gandhi et al., 2015).

Our study showed that recent fish Hg levels in coolwater and warmwater predators have increased in a majority of secondary watersheds and changes in climate may be an important driver in Ontario freshwater ecosystems. This investigation is one of few studies conducted on a large subset of the Ontario quarter million inland lakes. With rising temperatures, altered precipitation events, changing global climate indices and possible increases in global Hg emissions, predicting how and why fish Hg will change in the next few decades will be both vital and challenging. This is particularly demanding because fish Hg across a region may respond variably. In the Laurentian Great Lakes, for example, fish Hg levels have been decreasing in Lakes Ontario and Huron but increasing in Lake Erie due to factors such as recycling of historical releases and changes in food web structures (Bhavsar et al., 2010). For inland lakes, Hg levels in Ontario top predator fish have been increasing in recent years (Gandhi et al., 2014), particularly in northern Ontario. Gandhi et al. (2014) discussed how factors, such as Hg emissions from Asian countries, climate change, invasive species, and acidity of a lake, could be contributing to such fish Hg increases. With all these variables contributing to Hg levels, we need further understanding of (1) how Hg concentrations vary with Hg deposition, methylation, and uptake by living organisms; (2) the relationship between methylation rates and climatic factors; and (3) the key processes related to cycling of Hg and global transport (UNEP, 2013). To help assess temporal and spatial patterns associated with fish Hg levels, it is necessary to consider the concentration of atmospheric oxidants as they can affect the residency time and lead to changes in the deposition of Hg (Gustin et al., 2016). In addition, continued improvement and standardization of data collection and management and the implementation of biomonitoring procedures tailored to specific taxa is necessary to lessen the uncertainty of models and establish effective policies (Gustin et al., 2016). By acquiring improved data on Hg distribution and concentrations internationally, modelling capabilities under various climate models also advances (UNEP, 2013; Gandhi et al., 2015). As international efforts continue to limit and reduce current atmospheric Hg releases (UNEP, 2013), fish consumption of both wildlife and humans may be improved if greenhouse gas emissions are mitigated.

Acknowledgements

We thank MOECC for fish Hg data, Samantha Stefanoff, Thomas Van Zuiden, Katrina Gaibisels, Bailey Hewitt and Saudia Khan of York University for help in drafting the manuscript, Alessandro Filazzola for assisting with graphic design, Sogee Spinner for support in R-code development, in addition to 3 anonymous reviewers and Dr. Jose Domingo whose reviews greatly improved the manuscript. Funding for this study was provided from a Natural Science and Engineering Research Council Discovery Grant to SS.

Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.envres.2017.12.018.

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